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Inventory of aerosol and sulphur dioxide emissions from India: I—Fossil fuel combustion

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Abstract

A comprehensive, spatially resolved ($0.25° \times 0.25°$) fossil fuel consumption database and emissions inventory was constructed, for India, for the first time. Emissions of sulphur dioxide and aerosol chemical constituents were estimated for 1996–1997 and extrapolated to the Indian Ocean Experiment (INDOEX) study period (1998–1999). District level consumption of coal/lignite, petroleum and natural gas in power plants, industrial, transportation and domestic sectors was 9411 PJ, with major contributions from coal (54%) followed by diesel (18%). Emission factors for various pollutants were derived using India specific fuel characteristics and information on combustion/air pollution control technologies for the power and industrial sectors. Domestic and transportation emission factors, appropriate for Indian source characteristics, were compiled from literature. SO_2 emissions from fossil fuel combustion for 1996–1997 were $4.0\,Tg\ SO_2\,yr^{-1}$, with 756 large point sources (e.g. utilities, iron and steel, fertilisers, cement, refineries and petrochemicals and non-ferrous metals), accounting for 62%. $PM_{2.5}$ emitted was 0.5 and $2.0\,Tg\,yr^{-1}$ for the 100% and the 50% control scenario, respectively, applied to coal burning in the power and industrial sectors. Coal combustion was the major source of $PM_{2.5}$ (92%) primarily consisting of fly ash, accounting for 98% of the "inorganic fraction" emissions (difference between $PM_{2.5}$ and black carbon + organic matter) of $1.6\,Tg\,yr^{-1}$. Black carbon emissions were estimated at $0.1\,Tg\,yr^{-1}$, with 58% from diesel transport, and organic matter emissions at $0.3\,Tg\,yr^{-1}$, with 48% from brick-kilns. Fossil fuel consumption and emissions peaked at the large point industrial sources and 22 cities, with elevated area fluxes in northern and western India. The spatial resolution of this inventory makes it suitable for regional-scale aerosol-climate studies. These results are compared to previous studies and differences discussed. Measurements of emission factors for Indian sources are needed to further refine these estimates.

Keywords: SO_2; $PM_{2.5}$; Carbonaceous aerosols; Fly ash; Power plants; Brick-kilns; INDOEX

1. Introduction

It is now recognised that aerosols and sulphur dioxide (precursor gas to sulphate aerosol) play an important role in regional and global climate change, with some aerosol constituents (e.g. sulphate, organic matter (OM) and mineral matter) resulting in cooling and others (e.g. black carbon (BC)) in warming of Earth's atmosphere (Charlson et al., 1991, 1992; Liousse et al., 1996; Tegen et al., 1997). In contrast to greenhouse gases, aerosols have short atmospheric lifetimes (about few days to a week), and would concentrate in source regions, having climatic effects with strong spatial and temporal variations. Aerosol emissions must be accurately estimated, with good spatial resolution, as a first step to understand their transport and climatic effects, on a regional scale.

India is one of the fastest growing economies in Asia, with an annual average GDP growth of 6.1% (WDR, 2000). This has resulted in an increase in commercial energy consumption in the last decade (38% between 1990 and 1998) (CMIE, 1999; TEDDY, 1999). As there

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are no current estimates of fossil fuel consumption and pollutant emissions compiled by Indian regulatory agencies, these are needed for recent base years, with detailed spatial resolution. This work is also relevant in context of the recently completed Indian Ocean Experiment (INDOEX) (1998–1999), to study the aerosol transport, chemistry, and their climatic effects over Indian subcontinent and Indian Ocean (Ramanathan et al., 1997). The objective of this work was to develop a comprehensive spatially resolved aerosols and SO_2 emissions inventory for India for the latest base year, to serve as an input to the aerosol-climate modelling studies related to INDOEX.

Previous global emissions inventories have included SO_2 and aerosol emissions from fossil fuel combustion in India (Cooke et al., 1999; Cooke and Wilson, 1996; Liousse et al., 1996; Penner et al., 1993; Spiro et al., 1992). In these inventories, national average estimates of per capita fuel consumption were used, which were reported, for example, by the International Energy Agency (IEA), for base years 1984–1990, and distributed based on population density. To improve the spatial distribution of the fuel use and emission estimates, some sectorisation was introduced to differentiate the power sector from other industries (Arndt et al., 1997; Akimoto and Narita, 1994). Further sectorisation was recently introduced (Garg et al., 2001) using fuel consumption data from Government of India databases for the base years 1990 and 1995.

In the previous estimates, emission factors of pollutants (SO_2, BC, OM) have been used at an aggregate level. Average fuel-based emission factors have been used, in general, without information about the production and pollution control technologies used in various industrial sectors. These would introduce uncertainties in the emissions, e.g. the SO_2 emitted or retained in a process, or the relative amounts of organic and elemental carbon formed at different combustion temperatures. Also, in the absence of technology related information, assumptions have been made to enhance pollutant emission factors by an arbitrary value (e.g. Cooke et al., 1999) above that applicable to the industrial process in a developed country. In many cases, global average fuel characteristics were used while, for example, Indian coals would have significantly higher ash and lower sulphur content than the global average (Coal Atlas of India, 1993).

In this study, an attempt has been made to develop a comprehensive emission inventory, which links plant-by-plant process and control technology, and appropriate fuel composition, to the best available emission factors to estimate emissions from different sectors. An analysis of the transportation and domestic sectors was made to identify and develop appropriate/realistic emission factors and estimate emissions for India. The objectives of the present paper are: (i) construction of fossil fuel consumption database for India for the base year 1996–1997 (the latest data available at the time this study was started), (ii) development of realistic emission factors, for SO_2 and aerosols, from industrial, transportation and domestic sources in India, (iii) construction of a spatially resolved ($0.25^\circ \times 0.25^\circ$) emission inventory for SO_2 and aerosols ($PM_{2.5}$ or particulate matter (PM) $<2.5\,\mu m$ diameter, BC, OM and the "inorganic fraction") and projection of the emissions to 1998–1999 (INDOEX period).

2. Method

India is divided into 25 states, which are administered through state governments and seven union territories (as of 1996–1997), directly administered by the union government. The fossil fuel consumption in each state varies considerably depending on industrial development, urbanisation and population. Each state is sub-divided into districts ($\sim$15–50) for micro-developmental planning (GoI, 1992) and the district is the lowest level, where energy consumption estimates can be obtained. At the start of this study, the latest fuel consumption data was available for 1996–1997, which was chosen as the base year. Projections for future years can be made, for example, as shown for 1998–1999 for the INDOEX period. Fossil fuel consumption was divided into four major sectors namely electric utilities (power generation), industrial, domestic and transportation (Fig. 1). The industrial sector was sub-divided into large point sources (LPS) (energy intensive industries including iron and steel, fertilisers, cement, refineries and petrochemicals and non-ferrous metals) and "others", and the transport sector was sub-divided into road and rail transport. Sectoral national consumption of coal, petroleum fuels and natural gas and state wise consumption of each fuel were derived from MoPN (1998), MoC (1997) and CMIE (1999). The plant wise consumption of various fuels for LPS was derived from reports of various industrial organisations (CMA, 1999; SAIL, 1998; FAI, 1998) and ministries of the Government of India (MoI, 1998; MoPN, 1998; CBIP, 1997). Where plant wise fuel consumption data was not reported, national fuel consumption in that sector was apportioned to each plant, using production as a proxy. "Others" industries were considered as non-point sources and state level fuel consumption was estimated using state wise production as a surrogate. Fuel consumption in the transportation and domestic sectors was estimated at the state level. Source and fuel-specific emission factors of SO_2 and aerosol constituents were used to estimate the emissions from each source and were spatially distributed to a grid size of $0.25^\circ \times 0.25^\circ$ ($\sim 25 \times 25$ km) (Fig. 1).

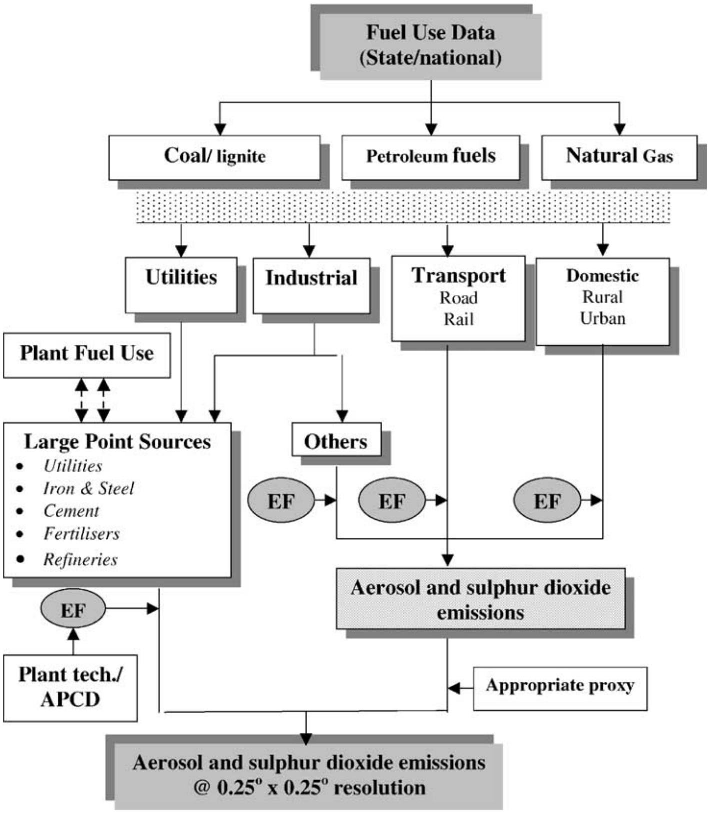


Fig. 1. Methodology for constructing a spatially resolved aerosol and sulphur dioxide emissions inventory from fossil fuel combustion.

3. Fuel consumption data

As part of this inventory, a fossil fuel consumption database was constructed for India for 1996–1997. This includes coal, lignite, petroleum fuels and natural gas consumption in utilities, industrial, domestic and transportation sectors.

3.1. Electric utilities (power generation)

In India, majority of the electric power is generated from coal-based thermal power plants with small amounts from gas- and oil-based plants. The total electricity generated during 1996–1997 from thermal power plants was 317×10^3 GW h, with coal accounting 88%, gas—9% and oil—3% (Vidyut Bharati, 1997). In thermal power plants, pulverised coal (PC) is used as the primary fuel and oil as the secondary fuel. The installed capacities and annual coal consumption at 100% load are reported by CBIP (1997). Using plant wise electricity generated (power load factors) during 1996–1997 (PIH, 1999; Vidyut Bharati, 1997), individual plant coal consumption was estimated. Plant wise oil consumption was estimated from specific oil consumption (kg $(\text{kW h})^{-1}$) (CEA, 1997) and electricity generated for each plant. Fuel consumption in gas- and oil-based power plants was estimated using installed capacities, respective power load factors for 1996–1997 (CEA, 1997; Vidyut Bharati, 1997) and specific fuel consumption values (CEA, 1997; MoPN, 1998). Wherever plant specific data on power load factors or specific oil consumption was not available, respective state average values were applied.

3.2. Industrial

Captive power plants: All major industrial units in India have power-generating units in their premises to meet their partial or full power requirement. The plant wise fuel consumption in captive power plants was estimated using installed generating capacities (PLR, 2000), average power load factors for different industrial sectors for each fuel type (CMIE, 1999) and specific fuel consumption (kg $(\text{kW h})^{-1}$). The specific fuel consumption values used were 0.90, 0.26 and 0.32 kg $(\text{kW h})^{-1}$ for coal, oil and natural gas, respectively. These values are the average for small electric utilities, which have similar capacities and technology.

Iron and steel plants, and coke ovens: There are seven integrated steel plants (ISPs) in India with a total production of 18.8 MT during 1996–1997. Apart from ISPs, the country produced 1.8 MT of iron and steel from secondary producers, 5.0 MT of sponge iron and 1.0 MT of alloy steel (SAIL, 1998) during 1996–1997. Coal is the major fuel consumed in iron and steel production along with small quantities of oil and gaseous fuels. SAIL (1998) reports fuel consumption in the ISPs. Fuel consumption in the secondary iron and steel, sponge iron and alloy steel plants was estimated using specific fuel consumption per ton of hot metal produced (SAIL, 1998; Das and Kandpal, 1997a) and production data. The difference between the reported national fuel consumption in the iron and steel sector and the fuel consumption accounted in the above plants (about 10%), was assumed to be consumed by non-specific sources and distributed to steel producing states in proportion to 1996–1997 steel production. The production of derived fuels (coke, COG, blast furnace gas) at ISPs were estimated using reported specific yield rates and at small coke ovens were estimated using ISPs average yield rates and assumed to be consumed in the iron and steel sector only.

Fertilisers: Between, nitrogenous and phosphatic fertilisers, 90% of energy is consumed by nitrogenous fertilisers in the form of feedstock and fuel. Using specific feedstock consumption values (ton of fuel per ton of fertiliser) (Das and Kandpal, 1998; FAI, 1998), and plant wise production of nitrogenous fertilisers (FAI, 1998), fuel consumption as feedstock was estimated. The difference of total fuel consumption and sum of feedstock and fuel use in captive power was used to estimate thermal energy consumption, and distributed to each plant in proportion to production.

Cement: The fossil fuels required in the cement plants are for thermal energy generation in cement kilns and captive power generation (Das and Kandpal, 1997b). Thermal energy requirement in the cement kilns depends on process type (wet, semi-dry and dry), with wet process requiring 1.8 times more energy than dry process. The difference of total fuel consumption and fuel use in captive power was assumed to be used in the kiln and distributed in proportion to 1996–1997 cement production, considering the process type (CMA, 1999), and variations in energy requirements (Das and Kandpal, 1997b).

Refineries and petrochemicals: CMIE (1999) and MoPN (1998) report the production capacities and fuel consumption in refineries and petrochemical plants. Here, it was assumed that naphtha is used as feedstock and will not contribute to emissions.

Pulp and paper: The plant wise production of pulp and paper is reported by IPPTA (1999). The difference of total fuel consumption and fuel use in captive power generation was apportioned to each plant in proportion to the production (IPPTA, 1998; CIER, 2000). Because of large number of plants, the fuel consumption was aggregated at district level.

Non-ferrous metals: Using specific fuel consumption of different fuels in non-ferrous metals production and 1996–1997 production data, fuel consumption at each plant was estimated (MoI, 1998; Department of Mines, 1999, 2000).

Mining: District wise captive consumption of coal for mining was taken from the Coal Directory of India (MoC, 1997). In the mining sector, petroleum products are used as fuels in the heavy machinery and total consumption was distributed to each district in proportion to quantity of total minerals mined (coal and non-coal) (MOL, 1997a, b).

Other industries: Industries other than the above were grouped as "others", which includes textile, engineering, chemicals and allied, glass and ceramics, food and plantation industries. The total fuel consumption in the respective categories was apportioned to each state using state-wise production as a surrogate (CII, 1998; CMIE, 1999; CIER, 2000). Fuel consumption in unclassified industrial categories was distributed to each state in proportion to total industrial output (millions of rupees) (CII, 1998).

Brick-making: In India, bricks are manufactured in small brick-kilns, where coal is used as the primary fuel, along with locally available low-grade fuels (e.g. rice husk, wood, crop waste, etc.), which vary from region to region. Brick-making is an unorganised sector and it is difficult to estimate the exact production and fuel consumption. Based on field surveys and consultations with Brick Manufacturers Associations Maithel (2000) estimated state wise brick production and corresponding coal consumption, considering regional variations in the fuel mix.

3.3. Domestic

Fossil fuels used in the domestic sector in India are kerosene, LPG and coal. District wise consumption of kerosene in urban and rural areas was estimated separately, using state average per capita consumption and fraction of population using kerosene (NSS, 1996). Reported total consumption of LPG (MoPN, 1998) and coal (MoC, 1997) in the domestic sector were distributed to each district using urban and total population as a proxy, respectively (GoI, 1998).

3.4. Transportation

In this inventory, only road and rail transportation sectors were considered, while shipping and aviation were not included. Diesel oil (diesel) and petrol (motor gasoline) are major fuels used in road transport in India. National consumption of diesel in the road transport is reported by MoPN (1998). Using state wise diesel vehicle population (MoST, 1998) as a surrogate, state wise diesel consumption was estimated. While estimating state wise diesel consumption, diesel vehicles were classified into light-duty and heavy-duty vehicles, and variations in the fuel economy and passenger kilometres travelled were considered (Bose, 1998). MoPN (1998) reports state wise petrol consumption in the road transportation. There are 22 major urban areas in India, which account for 22% of diesel and 35% of petrol vehicle population. These cities have been identified in each state and fuel consumption was calculated in proportion to respective vehicular population.

Diesel and small amounts of kerosene, petrol and coal are used in rail locomotives for traction and in workshops for production and maintenance. MoR (1998) reports national gauge-wise consumption (broad, meter and narrow gauges) of all fuels, which was apportioned to states in proportion to respective gauge-wise railway track length. Within each state, districts, which have railway tracks, were identified and fuel consumption distributed in proportion to geographical area of the districts.

4. Emission factors

Measured emission factors for Indian sources are not available in literature. The strategy followed here was to identify emission factors formulations, which could be customised to fuel composition and technologies used in India, derived from Indian data sources.

4.1. Sulphur dioxide

Sulphur dioxide emissions from fuel combustion and energy conversion (coke making) were included in this inventory. SO_2 emission factors for fuel combustion are a function of fuel sulphur content, combustion technology and pollution control equipment employed. During coal combustion, some fraction of sulphur remains in the bottom ash and remaining is emitted into the atmosphere in the form of SO_2. We developed emission factors for fuel characteristics/combustion technology typical in Indian industrial sectors.

4.1.1. SO_2 from coal

The sulphur content of Indian coals ranges from 0.1% to 0.8%, with exception of Assam coals, which have a higher value of 3.9% (Coal Atlas of India, 1993). Thermal power plants account for 73% of total coal consumption (299 million tons) and estimated SO_2 emissions from India would be sensitive to the assumed coal sulphur content for this sector. To make realistic SO_2 emission estimates, reported plant wise coal sulphur content was used (CBIP, 1997), which ranged from 0.10% to 0.80%, with a consumption weighted average of 0.59%. For coal consumption in the cement sector, state wise coal sulphur content was calculated as average of feeder coal-fields (CMA, 1999). In case of captive consumption of coal, average mine wise sulphur content was applied.

For coal consumption in other industries, transportation and the domestic sector, sulphur content was derived in the following manner. For coal producing states, the state wise production weighted average was calculated from mine wise production data and respective sulphur contents (MoC, 1997; Coal Atlas of India, 1993). It was assumed that inter-state transport between coal producing states is negligible. National production based weighted average sulphur content was calculated and applied to the states, which do not produce coal (Table 1). In northeast region, Assam is the only state, which produces coal. Due to geographical location, Assam coals are not transported out of northeast region, and hence not included in the national average. Assam coal sulphur content was applied for all other northeast states, which are primary consumers (Table 1). Lignite is produced in Gujarat and Tamil Nadu, and it is consumed in the lignite-based power plants, brick-kilns and domestic sector. Here, it was assumed that lignite not transported out of these states, as the production was far lower than total coal consumption for these states. Indigenous coals account for 97% of total coal consumption in India (MoC, 1997), and the sulphur content of imported coals was also assumed to be same as Indian coals.

Table 1
State wise sulphur content of coal and lignite in India

State	S content (wt%)
Coal	
Andra Pradesh	0.56
Assam	3.95
Bihar	0.58
Jammu & Kashmir	0.65
Maharashtra	0.63
Madya Pradesh	0.44
Orissa	0.60
Uttar Pradesh	0.60
West Bengal	0.60
Northeast states[a]	3.95
All other states and UTs[b]	0.60
Lignite	
Gujarat	0.5
Tamil Nadu	3.0

[a] Assam coal sulphur content is assumed for all northeast states (Arunachal Pradesh, Meghalaya, Mizoram, Nagaland, Sikkim and Tripura).

[b] UTs: union territories.

For SO_2 emissions from coke production, plant wise coke and coke oven gas (COG) production rates, for ISPs, were compiled from SAIL (1998). In the conversion process of coal to coke, it was assumed that 62% of sulphur remains in the coke and 38% goes to COG (Kato and Akimoto, 1992). For small coke ovens, average of ISP's coke and COG conversions rates were applied to estimate coke and COG production, and resulting SO_2 emissions. Coking coal sulphur contents used were respective state averages and range 0.44–0.65% (Table 1).

For coal combustion in power plants, it was assumed that 2.5% of sulphur is retained in the bottom ash (Kato and Akimoto, 1992; Arndt et al., 1997). A plant-by-plant analysis of power plants revealed that only 1.2% of total coal is burned in boilers having flue gas desulphurisation devices (pollution control equipment) and we, therefore, assumed that SO_2 emissions are uncontrolled. The same assumptions were applied to coal combustion in the captive power plants, operated by different industries. For industrial and domestic coal combustion, the combustion temperatures are lower than in utilities, and this results in higher sulphur retention in the bottom ash. We assumed 22.5% and 40%, respectively, of sulphur retention in the bottom ash for industrial and domestic coal combustion (Kato and Akimoto, 1992; Arndt et al., 1997) and uncontrolled SO_2 emissions.

4.1.2. SO_2 from petroleum fuels and natural gas

The petroleum fuels sulphur content (0.2–4.0%) used here is representative of Indian oil refineries (IOCL, 2000; Garg et al., 2001) and assumed constant for all states (Table 2). Low sulphur diesel oil is used for road transport in the four metropolitan cities (Calcutta, Chennai, Delhi, Mumbai), and a value of 0.5% was applied for these cities. Natural gas sulphur content was set as 0.00064% (Kato and Akimoto, 1992). For petroleum fuels and natural gas combustion in all the sectors, it was assumed that 100% of sulphur in the fuel is released into the atmosphere (Kato and Akimoto, 1992; Arndt et al., 1997) and emissions are uncontrolled.

Table 2
Sulphur content of petroleum fuels and natural gas in India

Fuel	S content (wt%)
Petrol (gasoline)	0.2
High speed diesel oil (HSDO)[a]	1.0
Low speed diesel oil (LSDO)	1.8
Fuel oil (FO)	4.0
Low sulphur heavy stock (LSHS)	2.0
Kerosene	0.2
Liquefied petroleum gas (LPG)	0.02
Natural gas	0.00064

[a] For road transport in metropolitan cities (Calcutta, Chennai, Delhi and Mumbai)—0.5%.

4.2. $PM_{2.5}$ and chemical constituents

During the fossil fuel combustion, PM of size ranging from sub-micron to several orders of micrometers are emitted. However, larger particles settle within short period of time and only particles $<2.5\,\mu m$ have a atmospheric life time about one week would travel several hundred kilometres and have climatic effects. The PM emissions consist of carbonaceous fraction, inorganic mineral ash and trace elements. The carbonaceous aerosols are of two types of BC and OM. The PM emissions depend on the fuel characteristics and combustion technology (coal ash content, boiler type, vehicle type, etc.), whereas the carbonaceous aerosol fraction depends on the combustion technology/characteristics. The measured carbonaceous aerosols emission factors are for BC and organic carbon (OC), a OM to OC ratio of 1.3 was assumed to account for hydrogen, nitrogen and other species in the carbonaceous aerosols (Countess et al., 1981), for all combustion sources. The difference of $PM_{2.5}$ and carbonaceous aerosols (sum of BC and OM) was assumed as "inorganic fraction" and emission factors were derived for all sources.

4.2.1. Coal combustion

4.2.1.1. Utilities (power generation). In India, 73% of total coal is consumed in utilities and the high ash content of pulverised coals (18–50%, with a consumption weighted average of 39%), would make the estimated PM emissions very sensitive to the emission factors for this sector. In view of this, a plant-by-plant analysis of power plants was undertaken, to identify the boiler types and installed air pollution control devices (APCD). CBIP (1997) reports status of Indian thermal power plants for 1997, including fuel type and characteristics, boiler type and capacity, and installed APCD along with their efficiencies. PC fired dry and/or wet bottom boilers account 85%, and other boiler types 1% (cyclone furnace, spreader stoker, underfed/overfed stoker), with 14% not reporting. The reported installed APCD are electrostatic precipitators (ESP)—73%, ESP with combination of others—22%, multiple cyclones (MC)—3%, with 2% not reporting. The above percentages (%) are that of amount of coal burned in the specific boiler type and boilers equipped with APCD.

Size resolved PM emission factors are reported for coal combustion for a combination of boiler type and APCD as a function coal ash content (USEPA, 1996). The above emission factors were measured for coal burning boilers in the United States, with average APCD operating efficiencies of 99.2% for ESP, 80–94% for MC, respectively for total PM removal. To evaluate the suitability of the AP-42 emission factors

(USEPA, 1996), we compared them to emission factors calculated using the current Indian emissions standards. In India, under the Air (Prevention and Control of Pollution) Act—1981, the emission standard for suspended particulate matter (SPM) concentrations from coal boilers of capacity $>150\,\mathrm{T\,day^{-1}}$ capacity is $150\,\mathrm{mg\,Nm^{-3}}$ (CPCB, 1996). For the power plants utilities boiler capacities, coal-firing rate, coal characteristics (e.g. ash content) and installed APCD are reported by CBIP (1997). For the boiler capacities of 30, 60, 110, 210 and 500 MW, which are representative of Indian utilities coal boilers, using the reported flue-gas flow rate for these boilers (CBIP, 1997) and assuming the highest allowable emissions of $150\,\mathrm{mg\,Nm^{-3}}$, resulting emission factors of PM were calculated to be $0.46–0.71\,\mathrm{g\,kg^{-1}}$. The PM emission factors derived from USEPA AP-42 for the same boiler types, existing APCD (including electrostatic precipitators and multiple cyclones), respective coal ash content, were $1.0–1.4\,\mathrm{g\,kg^{-1}}$, assuming APCD functioning 100% of the time, a factor of 1.8 higher than maximum allowable emission factors. In the absence of data on APCD malfunctioning, we also derived emission factors for a scenario of APCD not in operation 50% of the time, which ranged $1.6–2.8\,\mathrm{g\,kg^{-1}}$ and were about 3.5 times higher than allowable emissions under the present regulations. Therefore, the AP-42 formulation for both 50% and 100% control scenarios resulted in conservative emission factors, which were 1.8–3.5 times higher than emissions, if current Indian emission standards were being met. This implies that the particulate emissions standards are unlikely to be met under most current operating conditions, but needs verification through actual stack measurements.

Using AP-42 PM emissions size resolution for various firing configurations (USEPA, 1996), PM $<2.5\,\mu\mathrm{m}$ emission factors were derived for each plant for boiler type and APCD, using respective coal ash content (CBIP, 1997). For plants where boiler types were not reported (14%), an average of mostly used boilers (dry and wet bottom) types along with installed APCD was assumed, and for plants where APCD was not reported, an average of possible APCD for the given boiler type was assumed. With the assumption of 50% of time APCD not in operation calculated $PM_{2.5}$ emission factors range from 1.1 to $19.0\,\mathrm{g\,kg^{-1}}$, with consumption weighted average of $8.1\,\mathrm{g\,kg^{-1}}$. Uncontrolled particulate emission factors for lignite combustion in dry bottom boilers are similar to PC combustion (USEPA, 1996), and we assumed that controlled emission factors from lignite would be the same as from PC for the respective APCD. The resultant average $PM_{2.5}$ emission factors were lower than from coal (Table 3), because of low ash content (10%) of Indian lignite, a factor of four less than that of coal (39%).

There is a paucity of directly measured emission factors of carbonaceous aerosols from coal combustion, especially for OC. However, there are reported measurements of BC and OC as a fraction of PM emissions (Shibaoka, 1986; Veranth et al., 2000; Henry and Knapp, 1980; Olmez et al., 1988; Fisher et al., 1978). We derived BC and OC emission factors as a fraction of PM, measured downstream of the APCD. Average total carbon (TC) and BC fraction of PM were given as 4.3% (2.2–6.4%) (Shibaoka, 1986; Veranth et al., 2000; Henry and Knap, 1980) and 0.95% (0.9–1.0%) (Olmez et al., 1988; Fisher et al., 1978), respectively. The difference of TC and BC was given to be OC (average of 3.1%) (Hangebrauck et al., 1964). BC and OM emission factors calculated from above BC and OC fractions, and the $PM_{2.5}$ emission factors in the previous section

Table 3
$PM_{2.5}$, black carbon and organic matter emission factors for coal and lignite combustion (50% control scenario)

Source	Emission factor ($\mathrm{g\,kg^{-1}}$)		
	$PM_{2.5}$	Black carbon	Organic matter[a]
Coal			
Utilities (power generation)[b]	8.10	0.077	0.33
	(1.10–19.00)	(0.01–0.18)	(0.044–0.77)
Industrial boilers, captive power generation[c]	1.36	0.0095	0.040
	(1.19–1.42)	(0.0083–0.0099)	(0.035–0.042)
Domestic, brick-kilns, rail locomotives[d]	12.20	1.83	10.15
Lignite			
Utilities (power generation)[b]	2.04	—	0.19
	(1.86–3.91)		(0.18–0.38)
Domestic, brick-kilns[d]	4.60	0.18	3.89

[a] OM/OC ratio assumed as 1.3 (Countess et al., 1981).
[b] Consumption weighted average of plant wise values.
[c] Consumption weighted average of state wise values.
[d] Uncontrolled.

were 0.077 (0.01–0.18) and 0.33 g kg^{-1} (0.044–0.77), respectively (Table 3). Lignite combustion in the power plants has been seen to result in negligible amounts of BC emissions, while OC was about 7.5% of the emitted $PM_{2.5}$ (Pinto et al., 1998). This resulted in an OM emission factor of 0.19 g kg^{-1} (0.18–0.38 g kg^{-1}).

4.2.1.2. Industrial. High calorific content and low ash coals are used in industrial boilers in India. The commonly used industrial boilers are cyclone furnace, spreader stoker, underfed/overfed stokers, with APCD multiple cyclones, ESP and baghouse. In absence of sector specific data on type of boilers used, an average uncontrolled $PM_{2.5}$ emission factors for above boilers was calculated from USEPA AP-42 (USEPA, 1996). The ratio of controlled to uncontrolled emissions for above boiler types and APCD is 0.5, which was used to estimate controlled $PM_{2.5}$ emission factors. While calculating $PM_{2.5}$ emission factors, for coal producing states, the state wise production weighted average coal ash content was used and for non-coal producing states, the national production weighted average ash content was applied. Once again, for the 50% control scenario $PM_{2.5}$ emission factors were 1.19–1.42 g kg^{-1}, with an average of 1.36 g kg^{-1}. As installed industrial captive power plant capacities are small (PLR, 2000), and boilers are similar to industrial boilers, $PM_{2.5}$ emission factors were assumed same as for industrial boilers. In the captive power plants at integrated iron and steel plants, PC is fired in large capacity dry/wet bottom boilers and emission factors used were the same as those for utilities. The lower average ash content of coal used in the industrial sector (23%) resulted in average $PM_{2.5}$ emission factor a factor of six lower than for utilities, using higher ash PC (39%).

BC and OC emission factors, once again, have been derived from $PM_{2.5}$, using reported BC and OC fraction of particulate matter. In the absence of other information, for the industrial sector we use the TC/PM ratio of 0.1 used by Streets et al. (2001) for China and BC/OC ratio same as for the power sector. The resultant BC and OM emisison factors are 0.0095 (0.0083–0.0099) and 0.040 g kg^{-1} (0.035–0.042), respectively.

4.2.1.3. Domestic. Lower combustion temperatures in domestic coal combustion result in higher PM emissions, because of incomplete combustion of fuel. The measured PM emission factors for domestic coal combustion reported in literature range 7.6–31.0 g kg^{-1}, with an average 13.6 g kg^{-1} (Mumford et al. 1987; Butcher and Ellenbecker, 1982; Jaasma and Macumber, 1982; Hangebrauck et al., 1964; Sanborn, 1982; Truesdale and Cleland, 1982; Bond, 2000), and 90% of mass lies $<2.5\,\mu m$ (Macumber and Jaasma, 1982). Assuming emissions are uncontrolled, this gives a $PM_{2.5}$ emission factor of 12.2 g kg^{-1} (Table 3). The measured average $PM_{2.5}$ emission factor for domestic lignite combustion was 4.6 g kg^{-1} (2.7–6.5 g kg^{-1}) (Bond, 2000). Measured BC and OC fractions of PM for domestic coal combustion were 15% and 64%, respectively (Mumford et al., 1987). Because of low combustion temperatures, the OC fraction is much higher than industrial boilers. Reported BC and OC fractions for domestic lignite combustion were 4% and 65%, respectively (Pinto et al., 1998).

About 4% of lumped coal is fired along with locally available low-grade biofuels in brick-kilns (Maithel, 2000). The combustion temperatures are similar to domestic coal combustion and emissions are uncontrolled, in the absence of measured emission factors ($PM_{2.5}$, BC, OC) for brick-kilns, we used reported domestic emission factors. In Gujarat and Tamil Nadu, lignite is used as fuel in the brick-kilns, and domestic emission factors for lignite were applied.

4.2.1.4. Transportation. In rail locomotives a small quantity (0.05%) of coal is used as fuel, and once again, domestic $PM_{2.5}$, BC and OC emission factors were applied, which are suitable for the low temperature combustion.

4.2.2. Petroleum fuels and natural gas combustion

4.2.2.1. Utilities and industrial. PM emissions from petroleum fuel combustion are mostly in the smaller particle size range, with $PM_{2.1}$ mass accounting over 90% (Cass et al., 1982). $PM_{2.1}$, BC and OC emission factors for fuel oil, diesel oil and natural gas combustion in utilities and industrial boilers were reported by Cass et al. (1982) and Gray (1986), which were derived from reported measurements (Table 4). Bocola and Cirillo (1989) reports PM emission factor of 0.35 g kg^{-1} for industrial kerosene combustion. Kerosene is a fraction of distillate oil for which Gray (1986) showed that 96.4% of emitted PM was $<2.1\,\mu m$ diameter, with BC and OC constituting 8.5% and 5.5% of the PM, respectively.

4.2.2.2. Domestic. Gray (1986) reported $PM_{2.1}$, BC and OC emission factors for domestic LPG combustion. The measured average PM emission factor for Indian kerosene cookstoves was 1.95 g kg^{-1} (TERI, 1987) and using $PM_{2.1}$, BC and OC fractions as for industrial kerosene combustion, emission factors were estimated in this work (Table 4).

4.2.2.3. Transportation. There are no reported measurements of particulate or carbonaceous aerosol emission factors for Indian on-road vehicles. Here, we used an average of reported PM emission factors in other parts of World, likely to apply to Indian conditions. BC and OC emission factors were derived from average BC, OC fraction of PM and average PM emission factors. The

Table 4
$PM_{2.1}$, black carbon and organic matter emission factors for petroleum fuels and natural gas combustion in utilities, industrial and domestic sectors[a]

Source	Emission factor ($g\,kg^{-1}$)		
	$PM_{2.1}$	Black carbon	Organic matter
Utilities (power generation)			
Natural gas (boilers)	0.05	0.00	0.00
Natural gas (turbines)	0.27	0.00	0.02
Fuel oil	0.36	0.01	0.07
Diesel oil	0.67	0.06	0.06
Industrial			
Natural gas	0.34	0.00	0.03
LPG	0.31	0.00	0.03
Fuel oil	0.65	0.01	0.08
Diesel oil	0.97	0.08	0.08
Coke oven gas	0.23	0.00	0.02
Kerosene	0.34	0.03	0.03
Diesel oil (IC engines)	4.37	0.17	0.00
Petrol (IC engines)	1.03	0.02	0.24
Residential/commercial			
LPG	0.33	0.01	0.03
Kerosene	1.90	0.16	0.16

[a] Adapted from Cass et al. (1982), Gray (1986), Bocola and Cirillo (1989), TERI (1987).

average PM emission factor for heavy-duty diesel vehicles was taken as $4.62\,g\,kg^{-1}$, higher than light-duty diesel vehicles ($3.62\,g\,kg^{-1}$) (Table 5a). However, the carbonaceous aerosol fraction for light-duty vehicles is reported to be higher (95%) than that for heavy-duty vehicles (88%) (Table 5a). Leaded petrol vehicle PM emission factor of $0.84\,g\,kg^{-1}$, is higher than that for unleaded petrol vehicle without catalytic converters ($0.36\,g\,kg^{-1}$), with the latter having higher carbonaceous fraction (98%), than the former (67%) (Hildemann et al., 1991; Williams et al., 1989b; Szkarlat and Japar, 1983). To minimise the uncertainty introduced from using emission factors measured for non-Indian vehicles, we included last 20 years measurements (1981–2000), while calculating average emission factors, to represent the age distribution of Indian vehicles (Table 5a). Since over 95% of PM emissions from automobiles (diesel and petrol driven) are in the sub-micron size range (Williams et al., 1989a, b), we assumed all PM emissions from the vehicular sources are $<2.5\,\mu m$.

In India, diesel oil is the major petroleum fuel used in rail locomotives, along with small quantities of kerosene and petrol (MoR, 1998). USEPA (1997) reports a PM emission factor of $81.9\,kg\,(10^9\,BTU)^{-1}$ for diesel oil combustion in rail locomotives. Assuming same emission factor (kg per unit energy) for kerosene and petrol, using respective net calorific values, PM emission factors were calculated. $PM_{2.1}$, BC and OC fractions were assumed same as industrial combustion of respective fuels (Table 5b).

5. Fossil fuel consumption in India (1996–1997)

For comparison of consumption of various fuels, mass of fuels (kilo-tons) were converted into energy equivalents using respective net calorific values (MoPN, 1998; TEDDY, 1997). Total fossil fuel energy consumption in India during 1996–1997 was 9411 PJ. This includes fuel consumption for energy (83%) in the utilities, industrial, domestic and transport sectors, and feedstock/raw material (17%) in the industrial processes. The shipping and aviation sectors were not included in this study, but are likely to contribute $<1.5\%$. The highest sectoral consumption is in industrial (43%—all industrial categories), followed by utilities (37%), transportation (17%) and domestic (3%) (Fig. 2a). The majority of energy consumption in the utilities is from coal combustion, in case of transportation it is from diesel oil combustion. Coal/lignite account for 54% of the total energy, followed by diesel oil (18%), natural gas (12%) and other fuels (16%). Majority of the energy from coal is consumed in the utilities, whereas energy from diesel oil and natural gas are consumed in the road transportation and fertilisers sectors, respectively (Fig. 2a).

Five major states account for 56% of total energy consumption (Uttar Pradesh—14%; Maharashtra—13%; Madhya Pradesh—11%; Gujarat—10%; Andhra Pradesh—8%). In Uttar Pradesh, Madhya Pradesh and Andhra Pradesh, majority of the energy consumed is in the utilities (coal), whereas, in Maharashtra and Gujarat it is in the industrial and transportation sectors (coal and petroleum fuels). There are total 756 LPS, and account for 55% of total energy consumption resulting in energy consumption 5×10^3–$200 \times 10^3\,GJ\,km^{-2}$ in corresponding grids (Fig. 2b). Industrial clusters resulted in energy consumption of 1000–$5000\,GJ\,km^{-2}$, over north and parts of south India, Maharashtra and Gujarat states. The major urban areas, densely populated regions have moderate energy consumption of 500–$1000\,GJ\,km^{-2}$. Where as, energy consumption over central India, western Rajastan, Jammu & Kashmir, northeast states is lowest because of absence of industrial and transportation activities.

6. Pollutant emissions

6.1. Sulphur dioxide emissions

The estimated SO_2 emissions from fossil fuel combustion for India for 1996–1997 are $4.03\,Tg\,SO_2\,yr^{-1}$. The

Table 5

(a) *Summary of particulate matter, black carbon and organic matter emission factors for road transportation*[a]

Fuel/vehicle type	Emission factors PM ($g\,kg^{-1}$)	Avg. PM ($g\,kg^{-1}$)	BC (% of PM)	Avg. BC (% of PM)	OC (% of PM)	Avg. OC (% of PM)
Diesel/heavy-duty vehicles	6.7[b], 6.6[c], 4.0[d], 2.6[e], 4.2[f], 2.2[g], 6.2[h]	4.62	33[b], 52[f], 41[g]	42 (1.93)[i]	45[b], 30[f], 3[g]	36 (1.66)[i]
Diesel/light-duty vehicles	3.9[b], 3.3[b], 3.8[c], 3.1[h], 3.1[j], 4.4[k], 3.9[k]	3.63	53[b], 64[b], 77[k], 72[k], 60[l], 83[m], 58[n]	67 (2.42)	16[b], 30[b], 20[k], 34[k], 60[l], 17[m], 34[n], 21[o], 17[o]	24 (0.86)
Leaded petrol	0.88[g], 0.77[p], 0.88[q]	0.84	8.1[g], 4.2[p]	6 (0.05)	65.5[g], 27.8[p]	47 (0.39)
Unleaded petrol (without catalytic converters)	0.36[p]	0.36	23[p]	23 (0.08)	73[p]	73 (0.26)

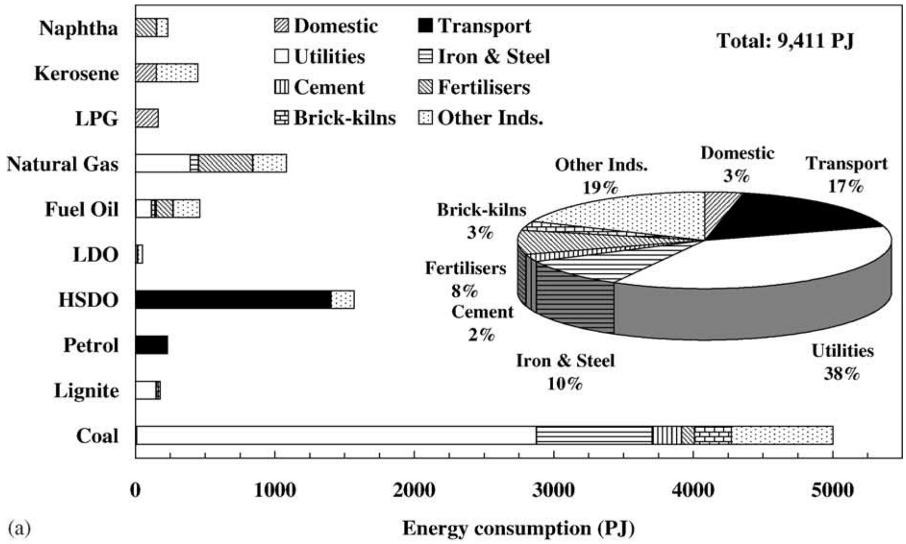
(b) *$PM_{2.1}$, black carbon and organic matter emission factors for petroleum fuels combustion in rail locomotives*

Fuel	Emission factors ($g\,kg^{-1}$) $PM_{2.1}$	Black carbon	Organic matter
Diesel oil	3.38	0.29	0.28
Petrol	3.59	0.07	0.65
Kerosene	3.33	0.29	0.29
Fuel oil	2.47	0.06	0.29

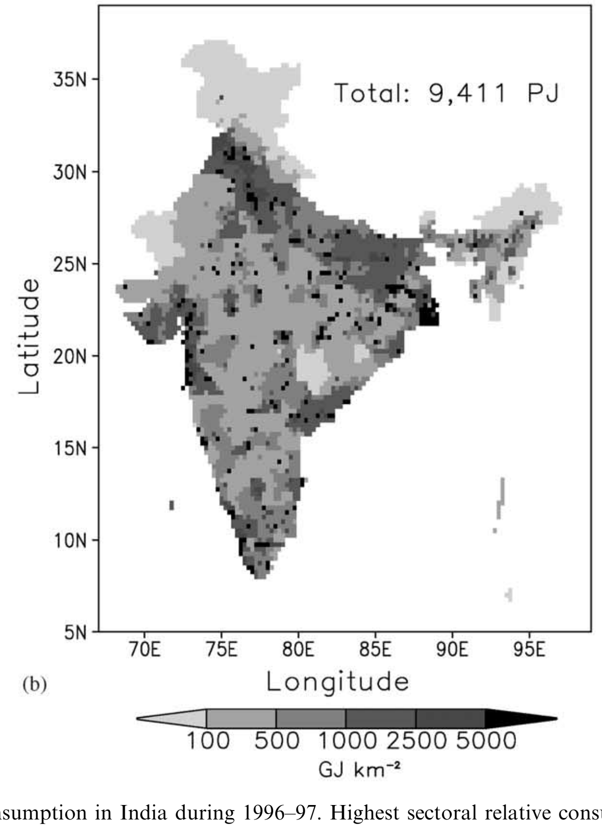
[a] PM: particulate matter, BC: Black carbon, OC: Organic carbon.
[b] Williams et al. (1989a).
[c] Wolff et al. (1981).
[d] Wang et al. (1997).
[e] Dietzmann et al. (1980).
[f] Lowenthal et al. (1994).
[g] Hildemann et al. (1991).
[h] Westerholm and Egebäck (1994).
[i] Values in the parenthesis are average BC and OC emission factor in $g\,kg^{-1}$.
[j] Bond (2000).
[k] Muhlbaier and Williams (1982).
[l] Postulka and Lies (1981).
[m] Bergin (1983).
[n] Frey and Corn (1967).
[o] Hammerle et al. (1994).
[p] Williams et al. (1989b).
[q] Szkarlat and Japar (1983).

highest sectoral relative contribution to SO_2 emissions is from utilities (50%), followed by industrial (35%) and transportation (15%) (Fig. 3a). The SO_2 emissions from utilities are primarily from coal combustion, and in transportation primarily from diesel combustion in road transport. Within industrial emissions, iron and steel and fertilisers together account only 10% of SO_2 emissions, because majority of the fuel is consumed as feedstock/raw material and not burnt as fuel. Coal/lignite account for 61% of emissions, followed by fuel oil/LSHS (19%), diesel oil (18%) and other fuels (2%). Significant contributions to SO_2 emissions are from coal, because of its high contribution of 54% to total energy consumption, and from fuel oil/LSHS from its high sulphur content (fuel oil—4%; LSHS—2%). While, natural gas share of energy consumption was 12%, because of lower sulphur content (0.00064%), SO_2 emissions are negligible (0.2%). India's power sector is primarily growing on coal with its total coal consumption almost doubling between 1989–1990 and 1997–1998. The increase in the coal consumption in the power sector would result in a proportionate increase in SO_2 emissions, if control measures are not implemented. LPS account for 62% of SO_2 emissions, and elevated release of SO_2 may have greater regional effects.

Highest SO_2 emission fluxes ($>5000\,kg\,SO_2\,km^{-2}$) are observed at grids in which power plants and other LPS are located (Fig. 3b). Higher emission fluxes are located (500–$5000\,kg\,SO_2\,km^{-2}$) in Tamil Nadu, Maharashtra, Uttar Pradesh, Gujarat, and West Bengal, as these states



(a)



(b)

Fig. 2. (a) Fossil fuel energy consumption in India during 1996–97. Highest sectoral relative consumption is in utilities followed by transportation and iron and steel. (b) Spatial distribution of fossil fuel energy consumption in India during 1996–1997.

together account for 52% of total emissions. Industrial clusters and major cities result in regional hot spots, in respective states. Emission fluxes in central and northwest India, are in the lower range (250–500 kg SO_2 km^{-2}), because of lower levels of industrial activity. Northeast states, Jammu & Kashmir and western Rajastan, experience very low emissions (0–100 kg $SO_2\,km^{-2}$), because of low industrial and transportation

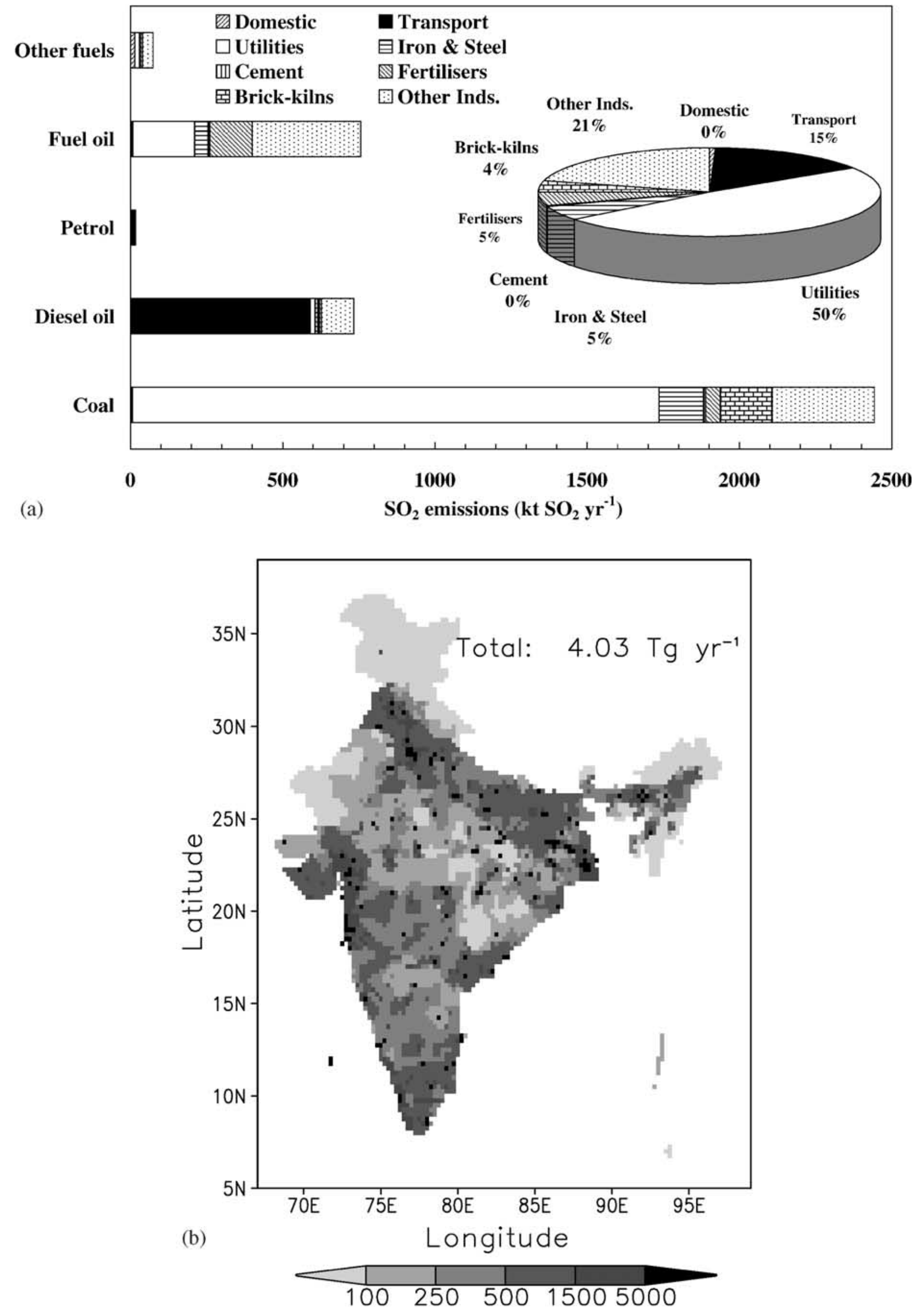


Fig. 3. (a) Sectoral relative contribution to SO_2 emissions (4.03 Tg yr^{-1}) is highest from utilities followed by transportation and "other industries" with coal combustion being the single largest contributor. (b) Spatial distribution of SO_2 emissions from fossil fuel combustion.

activity. However, use of high sulphur coals in this region resulted in moderate emissions over central Assam (100–250 kg SO_2 km^{-2}).

The present SO_2 emission estimates are a factor of 1.3 higher than those of Venkataraman et al. (1999) for 1990, resulting from increase in fuel consumption between 1990 and 1996–1997. Arndt et al. (1997) estimated 4.20 Tg SO_2 yr^{-1} for 1987–1988, accounting increase in fossil fuel consumption between 1987–1988 and 1996–1997 (Coal—70%; diesel oil—100%; fuel oil—30%), these estimates would scale to 6.40 Tg SO_2 yr^{-1}, in comparison to our estimate of 4.03 Tg SO_2 yr^{-1}. The discrepancy between two estimates mainly comes from assumed higher constant coal sulphur content (1%) by Arndt et al. (1997) for all sources and states, in contrast to plant wise values for power plants (0.1–0.8%), and state averages for other sources (0.44–0.65%) calculated from mine wise coal production and sulphur content, used in the present estimate. Our estimate compares well with recent estimates of Garg et al. (2001). The differences between this estimate and that of Garg et al. (2001) are our assumption of process based sulphur retention in bottom ash in contrast with their assumption of 100% sulphur released, for coal combustion. Also, their inventory included process emissions (H_2SO_4 production and non-ferrous metals smelting), which are not included in this estimate.

Previous SO_2 emissions inventories for India, used a single value of coal sulphur content between 0.5% and 1.0% for all sources. In contrast, the present inventory uses plant specific values for utilities and state average values for other sources, estimated from mine wise production and sulphur contents resulting in a more realistic estimate of national SO_2 emissions, source contributions and regional distribution.

6.2. $PM_{2.5}$ and chemical constituents emissions

Estimates have been made for both the 100% and 50% control scenarios. The $PM_{2.5}$ emissions for 1996–1997 for 100% and 50% control scenarios are 0.49 and 2.00 Tg yr^{-1}, respectively (Fig. 4). There is a four-fold increase in $PM_{2.5}$ emissions in 50% control scenario as compared to 100% scenario, primarily resulting from increase in emissions from coal combustion in the utilities. In 100% control scenario, on average installed APCD in utilities removes 97% of $PM_{2.5}$ emissions, where as in 50% control scenario removal is only 48%. However, the increase in carbonaceous aerosol emissions is only 10–15%, because coal combustion in the utilities is a minor source of these aerosols. The "inorganic fraction" of $PM_{2.5}$ emissions increases by a factor of 8.3 in the 50% control case, as $PM_{2.5}$ emissions from coal-based power plants primarily consist of "inorganic fraction" (96%). In the following sections all emission maps are for the 50% control scenario, which we believe is a conservative estimate of the emissions.

6.2.1. $PM_{2.5}$ emissions

$PM_{2.5}$ (particulate mass $<2.5\,\mu m$ diameter) emissions from fossil fuel combustion from India for 1996–1997 are 2.00 Tg yr^{-1} (Fig. 5). Majority of the $PM_{2.5}$ emissions (92%) are from coal combustion, followed by diesel oil (7%) and others (1%). The high ash content of PC (39%) results in these high emissions, in addition to

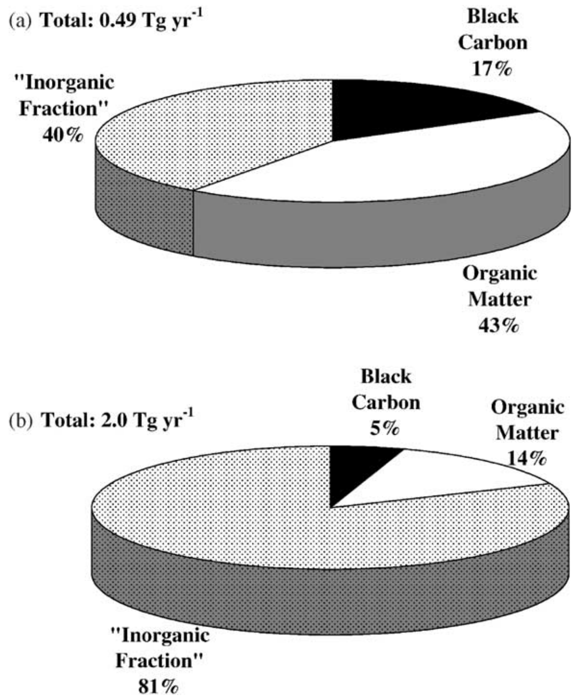


Fig. 4. $PM_{2.5}$ emissions from fossil fuel combustion in India, for 1996–97, for (a) 100% control scenario and (b) 50% control scenario (APCD operational only 50% of the time).

assumption of 50% control, which removes only 48% of $PM_{2.5}$ emissions. $PM_{2.5}$ emissions from diesel combustion are primarily from road transport. Combustion of fuels other than coal and diesel oil results in negligible amounts of $PM_{2.5}$ emissions. The sectoral relative contribution is highest from utilities (79%), followed by brick-kilns (8%) and transportation (7%). While, the amount of coal consumed in the brick-kilns is only 4% of the total, the firing practice of smouldering combustion leads to a high emission factor (12.2 g kg^{-1}), and results in higher emissions (8%). Heavy-duty diesel vehicles account 70% of total diesel oil consumption and relatively high emission factors result in 5% of total $PM_{2.5}$ emissions as compared to light-duty diesel vehicles (2%).

LPS result in emission fluxes >2000 kg km^{-2}, localised to the single $0.25^\circ \times 0.25^\circ$ (or 25×25 km) grid. High $PM_{2.5}$ emission fluxes (1000–2000 kg km^{-2}) are located over north India (Bihar, Uttar Pradesh, Punjab and Haryana), West Bengal, Maharashtra, Gujarat and Tamil Nadu. Higher emission fluxes in north India result from coal combustion in brick-kilns and higher densities of transportation and domestic sources. South, central, northwest India and central Assam have moderate emissions (50–250 kg km^{-2}) and western Rajastan and Jammu & Kashmir have the lowest emissions (<25 kg km^{-2}).

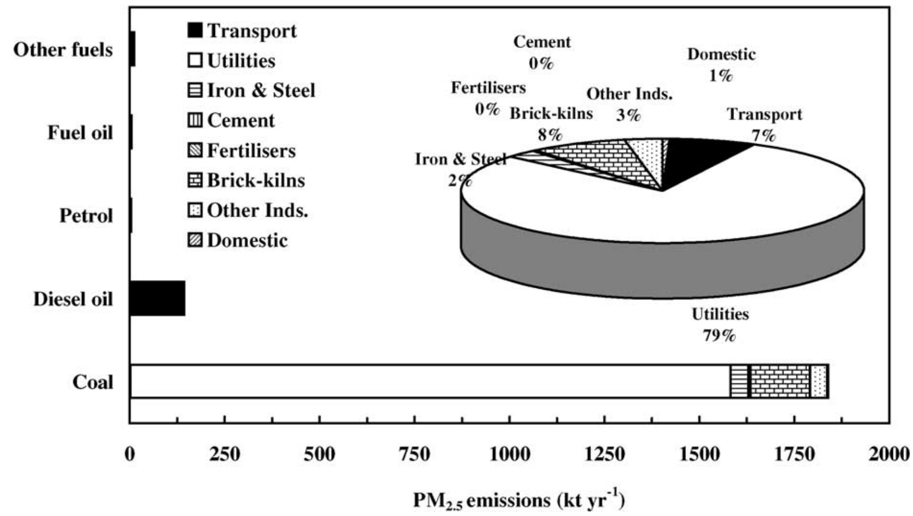


Fig. 5. Sectoral relative contribution to $PM_{2.5}$ emissions ($2.00\,Tg\,yr^{-1}$) is greatest from utilities, with coal combustion accounting 92% of total emissions.

Reported previous estimates of particulate emissions from India for 1990, include PM_{10} of $10.40\,Tg\,yr^{-1}$ (Wolf and Hidy, 1997) and $PM_{2.5}$ of $3.30\,Tg\,yr^{-1}$ (Reddy and Venkataraman, 1999, 2000). Present emissions are about a factor of 1.7 lower, if the previous emission estimates of Reddy and Venkataraman (2000) were projected for 1996–1997. This is because Reddy and Venkataraman (2000) assumed emissions from all sources to be uncontrolled, in comparison to the present inventory, which uses plant specific APCD for power plants and average possible APCD for other sources, for coal combustion. Other refinements in the present inventory include boiler/APCD specific emission factors for power plants, more realistic emission factors for other sources based on regional coal characteristics and source wise differentiation of emission factors for petroleum fuels combustion.

6.2.2. *Carbonaceous aerosol emissions*

BC emissions are $0.10\,Tg\,yr^{-1}$ from fossil fuel combustion from India for 1996–1997. Diesel oil and coal account for 59% and 40% of emissions, respectively. The major source of BC emissions is transportation (58%) followed by the brick-kilns (24%), utilities (15%) and others (3%) (Fig. 6). Although power plants and the industrial sector together account for 76% of primary energy consumption, because of high combustion temperatures and installed pollution control equipment, they result in only 18% BC emissions. Road transport accounts for only 16% of the energy consumption but contributes 57% of BC emissions, primarily from diesel combustion. Within the road transport sector the heavy-duty trucks account for more than 50% of fuel consumption and result in the 52% of BC emissions. Small-scale coal combustion including brick-kilns, rail locomotives, domestic and others, contribute to 23% of BC emissions. The low combustion temperature in small-scale coal combustion results in high PM emissions with a high carbon fraction, as reflected in the emission factors of $1.83\,g\,kg^{-1}$ (Table 3) chosen for these sources. The domestic sector accounts only 0.12% of the coal consumption in India (MoC, 1997) and results in 1% of BC emissions. This is in contrast to China, where domestic coal combustion accounts for about 80% of fossil fuel BC emissions (Streets et al., 2001).

Estimated OM emissions from fossil fuel combustion for 1996–1997 are $0.27\,Tg\,yr^{-1}$ (Fig. 7). The largest OM emissions are from coal combustion (75%) and with 24% from diesel oil and 1% other fuels. The highest relative contribution to OM emissions is from brick-kilns (48%) followed by utilities (24%) and transportation (25%). Once again lower combustion temperatures in the brick-kilns result in PM emissions with higher organic fraction. The use of diesel oil in road transport result 24% of OM emissions.

The four metropolitan cities (Calcutta, Chennai, Delhi and Mumbai) have the highest BC emission fluxes ($>500\,kg\,km^{-2}$) resulting from diesel combustion in the road transport sector. The grids of higher emissions are concentrated over major cities and industrial clusters and power plants ($150–500\,kg\,km^{-2}$). Industrial clusters result in moderate emissions ($50–150\,kg\,km^{-2}$) over Gujarat, Maharashtra, Tamil Nadu, West Bengal and Haryana, where as entire north India experience emissions of $25–50\,kg\,km^{-2}$ from coal combustion in brick-kilns. However, all other areas have emissions $<25\,kg\,km^{-2}$. The major source of OM emissions is coal

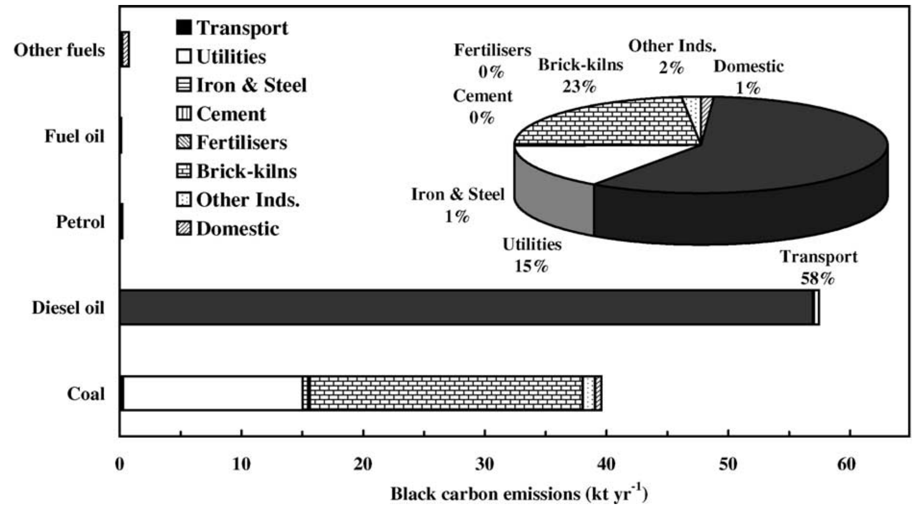


Fig. 6. Sectoral relative contribution to black carbon emissions ($0.10\,\mathrm{Tg\,yr^{-1}}$) is largest from diesel transportation.

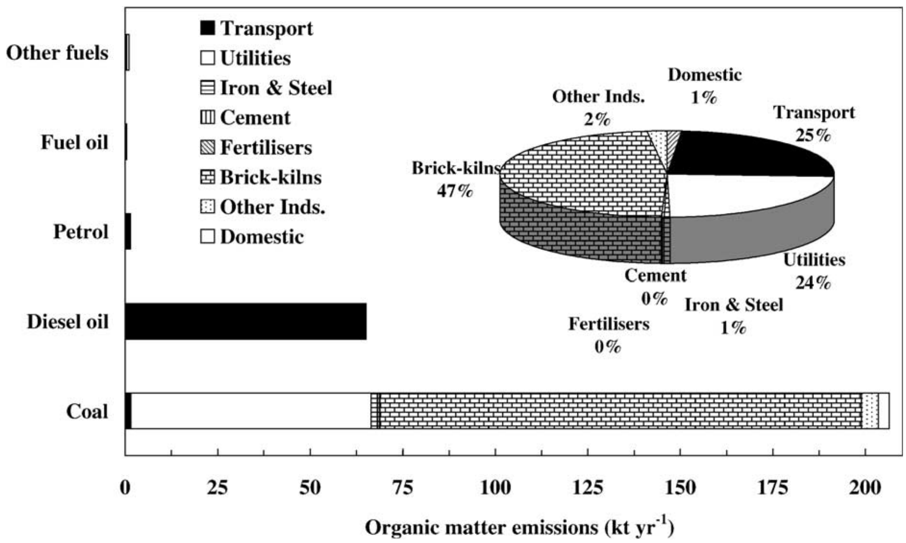


Fig. 7. Sectoral relative contribution to organic matter emissions ($0.27\,\mathrm{Tg\,yr^{-1}}$) is highest from brick-kilns followed by utilities and diesel transportation, with coal accounting for 77% of OM emissions.

combustion in brick-kilns results in high emission fluxes (100–$1000\,\mathrm{kg\,km^{-2}}$) in Bihar, Uttar Pradesh, Punjab and Haryana. Though OM fraction of $PM_{2.5}$ emissions from coal combustion in utilities is only 4%, the large coal consumption results in emission fluxes of $>1000\,\mathrm{kg\,km^{-2}}$ at all coal-based power plant locations.

In the recent global carbonaceous emissions inventory of Cooke et al. (1999) the BC emissions estimate of $0.31\,\mathrm{Tg\,yr^{-1}}$ for India, for 1984, is a factor of three higher than present estimates (Cooke, 1999). For coal combustion in the power/industrial sector Cooke et al. (1999) derived BC emission factors by assuming that 25% of sub-micron particulate emissions is BC for industrially developed countries, and enhanced them by a factor of five for developing countries (including India). For petroleum fuels combustion, also, average BC emission factors for industrial countries were enhanced by a factor of five to obtain BC emission factors for developing countries. The above assumptions resulted in higher BC emission factors for all sources, as compared to values used in the present study, yielded higher BC emissions. In addition, there are three global BC emission inventories (Penner et al., 1993; Cooke and Wilson, 1996; Liousse et al., 1996), which place global

BC emissions at 6.6–8.0 $Tg\,yr^{-1}$, with 1.9–2.5 $Tg\,yr^{-1}$ from Asia. However, country wise breakdown of emissions was not given to compare present estimates.

Reddy and Venkataraman (2000) had previously estimated uncontrolled BC emissions from fossil fuel consumption for India 0.25 $Tg\,yr^{-1}$, for 1990, which is about a factor of two higher than the current estimates. The higher BC emissions were resulted from use of very high BC emission factors (Cooke and Wilson, 1996), which had been obtained by equating post-APCD sub-micron particulate emissions to BC for coal combustion, and from non-differentiation of BC emission factors for petroleum fuels combustion in different sources. Refinements in emission factors since, include identification of combustion technology and coal characteristics, sectoral differentiation for coal and petroleum fuels combustion and derivation of BC emission factors as fraction of $PM_{2.5}$ or $PM_{2.1}$ emissions from reported measurements, lead to more realistic BC emissions in the present study.

Dickerson et al. (2001) estimated BC emissions from fossil fuel combustion from India at 0.04 $Tg\,yr^{-1}$, with an upper estimate (high case) of 0.36 $Tg\,yr^{-1}$. Our BC emission estimates compare well with the central values from this study and are somewhat more conservative for the transport and power sectors. In addition we include coal use in brick-kilns, a significant small-scale source and domestic coal use, as explained further. In their high case, Dickerson et al. (2001) assumed industrial coal to be combusted without any pollution control equipment, giving a 12.5 times larger BC emission factor, and high end BC emissions of 0.25 $Tg\,yr^{-1}$. We believe that the difference in the two emissions estimates result from differences in the sectoral coal use and the application of a high emission factor to a large fraction of the coal consumption. We are participating in studies to refine these emissions estimates using new emission factors and from general circulation model simulations for the INDOEX-IFP period, for evaluation against measurements.

The present OM emission estimate of 0.27 $Tg\,yr^{-1}$ is lower than previous estimates for India (Cooke et al., 1999; Reddy and Venkataraman, 2000). Cooke et al. (1999) estimated 0.30 $Tg\,yr^{-1}$ of OC emissions from India for 1984 (Cooke, 1999) and resultant OM emissions would be 0.39 $Tg\,yr^{-1}$ (using an OM/OC ratio of 1.3). Once again, the OC emission factors for all sources were enhanced by a factor of five for developing countries, resulting higher emissions. Reddy and Venkataraman (2000) derived OC emission factors assuming a constant OC/BC ratio of three for all sources and higher estimates of BC emissions resulted in higher OM emissions. In the present study, carbonaceous aerosols (BC and OC) emission factors for coal combustion, were derived as fraction of $PM_{2.5}$ for utilities and industrial sectors from reported measurements. These lie in the range of values reported by Bond (2000) and Bond et al. (1999) (BC: 3×10^{-3}–$1\times10^{-3}\,g\,kg^{-1}$; OC: 2×10^{-3}–$1\times10^{-2}\,g\,kg^{-1}$) from new measurements and a review of existing measurements. For all petroleum fuels sector/source wise emission factors were used. These refinements have resulted in the current estimates.

There have been recent assertions in the literature of the possible adulteration of petrol and gasoline/petrol in India (UNEP, 1999; Dickerson et al., 2001). This is believed to occur from the diversion of kerosene, subsidised for use as a domestic cooking fuel, for adulteration at the retail outlets (petrol pumps) of the distribution system. Indian oil refineries maintain fuel specifications (IS:2796-1995; IS:1460-1995) at the refinery, but the distribution system is not well monitored. While suitable methods for fuel testing (Patra and Misra, 2001) are being developed to establish the extent of this problem, it has not been possible to quantify the percentage of the petrol/diesel in India suffering adulteration at this time. In the high case of Dickerson et al. (2001), the BC emission factor for the transport sector was enhanced by a factor of 30 to account for poor maintenance of vehicles and possible adulteration of petrol/diesel with kerosene, leading to a high end estimate of 0.11 $Tg\,yr^{-1}$ from transport sector.

6.2.3. The "inorganic fraction" of $PM_{2.5}$

Aerosol chemical composition depends on the fuel type and combustion practices, with coal having a larger non-carbonaceous fraction compared to petroleum fuels. Especially high temperature coal combustion (e.g. utilities, industrial) results in PM emissions having a high inorganic fraction (Mohr et al., 1996) and a low carbonaceous fraction. The "inorganic fraction" from coal combustion primarily consists of fly ash particles, which predominates in the super-micron size range (McElroy et al., 1981) and small amounts of water soluble ions and trace metals. In this case, they were estimated as the difference between $PM_{2.5}$ and sum of "BC and OM" emissions.

The "inorganic fraction" emissions from India are 1.63 $Tg\,yr^{-1}$, for 1996–1997, with utilities alone contributing 92% of emissions (Fig. 8) and balance from industrial (5%) and other sources (3%). The large quantity of PC (73% of total consumption) with high ash content (39%) used in this sector, results in high $PM_{2.5}$ emissions, which primarily consist of "inorganic fraction" (94%). Though brick-kilns contributed 8% of $PM_{2.5}$ emissions, their contribution to "inorganic fraction" emissions is negligible, because the low combustion temperatures result primarily in carbonaceous aerosol emissions, with ash likely to remain as solid residue. Petroleum fuel combustion also results primarily in carbonaceous aerosol emissions, and their contribution to the "inorganic fraction" is negligible.

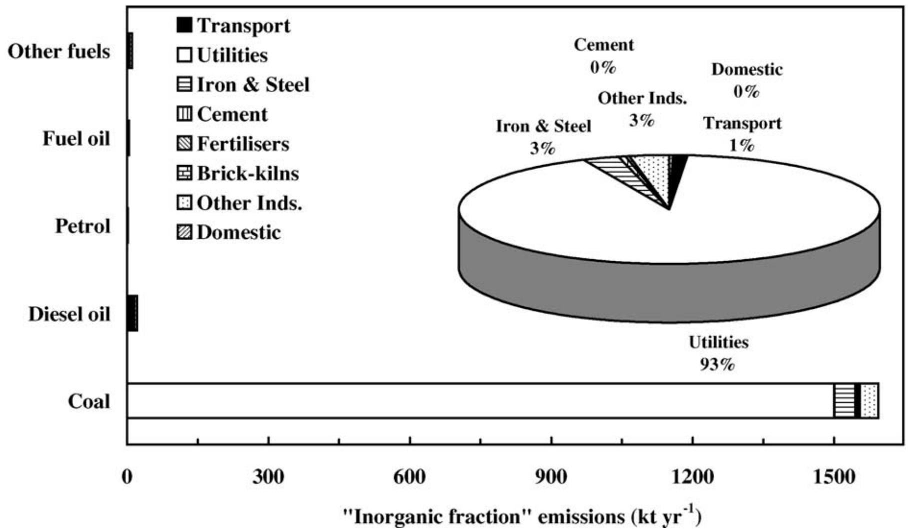


Fig. 8. Utilities are the primary source of "inorganic fraction" (i.e. fly ash) emissions (1.63 $Tg\,yr^{-1}$), with coal accounting 98% of the emissions.

The major source of "inorganic fraction" (i.e. fly ash) is coal combustion in power plants and other LPS, and results in highest emissions fluxes (1000–10000 $kg\,km^{-2}$). The domestic/brick-kilns coal combustion derived aerosols have a low "inorganic fraction", however higher source densities in north India results in moderate emissions fluxes. Non-specific sources of industrial combustion of coal result in moderate emission fluxes over industrial clusters in the states of Gujarat, Maharashtra, West Bengal, Tamil Nadu and Andhra Pradesh. Central, northwest India and Jammu & Kashmir have lower source densities and result in the lowest emission fluxes ($<25\,kg\,km^{-2}$).

For comparison, present "inorganic fraction" emissions a factor of 1.4 lower than the previous estimate of 2.24 $Tg\,yr^{-1}$ for India for 1990 (Reddy and Venkataraman, 2000). In the previous estimate emissions were assumed as uncontrolled, in comparison present study considers 50% control of emissions from coal combustion in utilities and industrial sectors, which are primary source of "inorganic fraction" emissions, leading to the lower estimate.

7. Extrapolation of pollutant emissions to 1998–1999 (INDOEX)

One of the primary objectives of present emissions inventory development is to serve as an input to the transport and climate modelling studies related to the recently completed INDOEX. Presently estimated SO_2 and aerosol chemical constituents emissions for 1996–1997 were extrapolated to 1998–1999 (Table 6), in proportion to increase in the fossil fuel consumption in utilities, industrial, domestic and transportation sectors. The sector wise fossil fuel consumption data from 1989–1990 to 1996–1997/1997–1998 was used to extrapolate the fuel consumption and resulting emissions for the 1998–1999 (April 1998–March 1999). Spatially resolved ($0.25^\circ \times 0.25^\circ$) SO_2 and $PM_{2.5}$ chemical constituents emission maps for INDOEX period can be constructed by multiplying the grid wise emissions for 1996–1997 and respective growth factors given in Table 6.

Table 6
SO_2 and aerosol emission projections for INDOEX period

Pollutant	Emissions ($Tg\,yr^{-1}$)		Growth factor
	1996–1997	1998–1999	
Sulphur dioxide	4.03	4.51	1.12
$PM_{2.5}$	2.00	2.20	1.10
Black carbon	0.10	0.12	1.14
Organic matter	0.27	0.28	1.06
"Inorganic fraction"	1.63	1.80	1.10
Total primary energy consumption (PJ)	9411	10409	1.11

8. Conclusions

A comprehensive, spatially resolved ($0.25^\circ \times 0.25^\circ$) fossil fuel consumption database and emissions inventory was constructed, for India, for the first time. Emissions of sulphur dioxide and aerosol chemical constituents were estimated for 1996–1997 and extrapolated to the Indian Ocean Experiment (INDOEX) study period (1998–1999), to serve as an input to aerosol-climate studies. A district level fossil fuel consumption database was developed, with sources including coal/lignite, petroleum fuels and natural gas consumption in power plants, industrial, transportation and domestic sectors. The fossil fuel energy consumption during 1996–1997 was 9411 PJ, with major contribution from coal (54%) followed by diesel (18%), natural gas (12%) and other fuels (16%). Coal consumption was primarily in the power sector, while diesel consumption was in road transportation. India has a coal-based power sector, with coal consumption almost doubling in this sector between 1989–1990 and 1997–1998, a trend which is likely to continue.

Emission factors for various pollutants were derived using India specific fuel characteristics and information on combustion/air pollution control technologies for the power and industrial sectors. Domestic and transportation emission factors, appropriate for Indian source characteristics, were compiled from literature. At present SO_2 emissions from all combustion sources are uncontrolled. The annual SO_2 emissions from fossil fuel combustion for 1996–1997 were 4.0 Tg $SO_2\,yr^{-1}$. There are 756 LPS, accounting for 62% of total SO_2 emissions. Coal combustion in the thermal power plants was the single largest contributor (43%) to SO_2 emissions, and balance emissions mostly from diesel use in road transport (18%) and fuel oil combustion in industrial sector (14%).

$PM_{2.5}$ emissions from fossil fuel combustion from India, for 1996–1997, were 0.49 and 2.0 Tg yr^{-1} for the 100% and the 50% control scenario, respectively, applied to coal burning in power plants and industrial sector. The four-fold increase in $PM_{2.5}$ emissions in the 50% control scenario is from use of high-ash Indian coals in the utilities. Power plant were the primary source of $PM_{2.5}$ emissions with a contribution of 79% followed by 8% from brick-kilns, and 7% from diesel transport. The $PM_{2.5}$ emissions from coal combustion primarily consist of "inorganic fraction" (i.e. fly ash particles), with power plants accounting 93% of "inorganic fraction" emissions of 1.6 Tg yr^{-1}.

BC and OM emissions from India, from fossil fuel combustion were only 29% and 22% of total emissions, respectively, with balance emissions from biomass combustion (*see companion paper*). Among fossil fuel sources, diesel use in road transportation was the single largest contributor to BC emissions accounting 58% of the total. Brick-kilns, specific to this region, are fired with coal and locally available biofuels and resulted in 48% and 24% of the OM and BC emissions, respectively. Diesel transport and coal fired power plants were other significant contributors to OM emissions.

The spatial resolution of this inventory makes it suitable for regional-scale aerosol-climate studies. The sector-specific emission factors for various pollutants were derived to closely represent Indian source emissions. Measurements of emission factors for Indian sources including sulphur retention rates in the boilers, operating efficiencies of air pollution control devices and characterisation of on-road vehicular emissions, are needed to further refine these estimates.

Note: Detailed tables of sector- and state-wise emissions, and emission maps of SO_2, $PM_{2.5}$, BC, OM and "Inorganic Fraction" are posted on *Aerosol Research Laboratory* website at http://www.iitb.ac.in/~cese/arl/eminv.htm.

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